



TITLE:

Ozonesonde observations at Christmas Island (2°N, 157°W) in the equatorial central Pacific

AUTHOR(S):

Takashima, H.; Shiotani, M.; Fujiwara, M.; Nishi, N.; Hasebe, F.

CITATION:

Takashima, H. ...[et al]. Ozonesonde observations at Christmas Island (2°N, 157°W) in the equatorial central Pacific. Journal of Geophysical Research: Atmospheres 2008, 113(D10): D10112.

ISSUE DATE:

2008-05-27

URL:

<http://hdl.handle.net/2433/217238>

RIGHT:

© 2008 American Geophysical Union. Further reproduction or electronic distribution is not permitted.

Ozonesonde observations at Christmas Island (2°N, 157°W) in the equatorial central Pacific

H. Takashima,^{1,2} M. Shiotani,¹ M. Fujiwara,³ N. Nishi,⁴ and F. Hasebe³

Received 11 September 2007; revised 10 January 2008; accepted 28 February 2008; published 30 May 2008.

[1] Ozone and water vapor observations have been conducted at Christmas/Kiritimati Island (2°N, 157°W) in the equatorial central Pacific as a part of the Soundings of Ozone and Water in the Equatorial Region (SOWER)/Pacific mission. We launched 33 ozonesondes and 33 chilled-mirror hygrometers in nine observation campaigns from 1999 to 2003 for various seasons. We found that ozone concentrations at Christmas Island are low in the whole troposphere (~10–35 ppbv), particularly in the marine boundary layer (MBL). Ozone variation is small throughout the year compared with other tropical stations, though annual and interannual variations of meteorological fields are large over the equatorial central Pacific. Just below the tropopause, during the August 2002 campaign, we observed substantially reduced ozone concentrations (<10 ppbv) similar to those found in the MBL, which are maintained at least for the observation period. From meteorological conditions, we found that air mass was advected from the Inter-Tropical Convergence Zone (ITCZ), located to the north of Christmas Island, in accordance with the northeasterly wind that is only observed during northern summer in the upper troposphere. The origin of air mass is supposed to be from the MBL in the ITCZ.

Citation: Takashima, H., M. Shiotani, M. Fujiwara, N. Nishi, and F. Hasebe (2008), Ozonesonde observations at Christmas Island (2°N, 157°W) in the equatorial central Pacific, *J. Geophys. Res.*, 113, D10112, doi:10.1029/2007JD009374.

1. Introduction

[2] Christmas (Kiritimati) Island, a part of the Republic of Kiribati, is located at 2°N and 157°W in the equatorial central Pacific. This is one of the most important regions in the equatorial Pacific to consider the east-west contrast of meteorological conditions associated with the Walker circulation and its variation due to the El Niño–Southern Oscillation (ENSO). However, atmospheric measurements including trace gas observations have not been extensively conducted around here, because there is no routine meteorological station. There was a series of ozone and water vapor sonde soundings conducted during the Central Equatorial Pacific Experiment (CEPEX) campaign in March 1993 [Vömel *et al.*, 1995; Kley *et al.*, 1996, 1997], but the space-time characteristics in the equatorial central Pacific have not been fully understood yet.

[3] Ozone and water vapor observations around the tropical tropopause are important from the view point of stratosphere-troposphere exchange (STE) processes which control the stratospheric air conditions. In particular, under-

standing of spatial and temporal structures in the tropical tropopause layer (TTL) is crucial [e.g., Folkins *et al.*, 1999, 2006; Kley *et al.*, 2000; Takashima and Shiotani, 2007]. Since the photochemical life time of ozone and water vapor is very long in the TTL, these gases can be used as a tracer. In addition, simultaneous observations of ozone and water vapor give us important information on the STE processes, because of the stratosphere-troposphere contrast of ozone and water vapor distributions with high ozone and low water vapor in the stratosphere and vice versa in the troposphere. Using ozone and water vapor data, we are able to further investigate the STE processes including the dehydration mechanism around the tropical tropopause as presented by Kley *et al.* [2000].

[4] As to a global-scale east-west contrast of the longitudinal ozone distribution in the tropical troposphere, the zonal wave number one structure with maxima around the Atlantic and Africa and minima around the western Pacific is well known [e.g., Fishman *et al.*, 1990; Shiotani, 1992]. The vertical characteristics of the zonal wave one structure were presented for the first time by Thompson *et al.* [2003] with ozonesonde data from the Southern Hemisphere Additional Ozonesondes (SHADOZ) archive. By paying special attention to the TTL, Takashima and Shiotani [2007] recently found that the TTL, wave one structure of ozone and the east-west temperature contrast in the upper troposphere could be explained by the large-scale atmospheric response to tropical heat source. However, there are still several weaknesses of the data coverage, because of only a few observations in the equatorial Pacific.

¹Research Institute for Sustainable Humanosphere, Kyoto University, Uji, Kyoto, Japan.

²Now at Atmospheric Composition Research Program, Frontier Research Center for Global Change, JAMSTEC, Yokohama, Japan.

³Faculty of Environmental Earth Science, Hokkaido University, Sapporo, Japan.

⁴Geophysical Institute, Kyoto University, Kyoto, Japan.

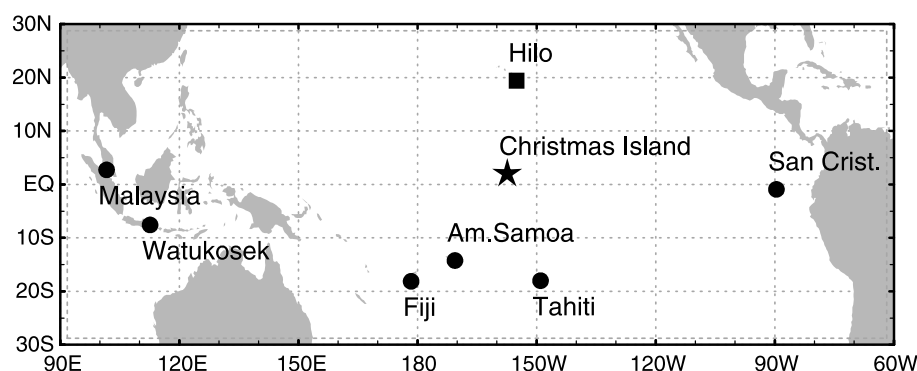


Figure 1. Locations of the tropical Pacific ozonesonde station from Southern Hemisphere Additional Ozonesondes (SHADOZ) (circles) and those of Hilo (Hawaii) and Christmas Island (157°W, 2°N) (star).

[5] Substantially reduced ozone concentration in the upper troposphere or just below the tropopause called “near-zero ozone concentration” is often observed in the tropical Pacific [e.g., Kley *et al.*, 1996; Solomon *et al.*, 2005]. A mechanism of the reduced ozone concentration is supposed to be due to vertical transport of low-ozone air from the marine boundary layer (MBL) by tropical deep convection as suggested by Kley *et al.* [1996]. As will be described further below, Solomon *et al.* [2005] recently investigated the spatial variation of the reduced ozone concentration in the tropics and subtropics using the SHADOZ ozonesonde data, and found high frequency of the reduced ozone in the equatorial Pacific.

[6] The Soundings of Ozone and Water in the Equatorial Region (SOWER)/Pacific mission has been running on a campaign basis since 1998 to improve our knowledge on ozone and water vapor distributions in the tropical Pacific by making coordinated radiosonde observations at several equatorial places. In addition to establishing climatology and variability in ozone and water vapor, the mission is intended to explore controlling dynamical/chemical processes for these species and to collect correlative observations for satellite data validation [e.g., Hasebe *et al.*, 2007]. See also the SOWER Web site (<http://sower.ees.hokudai.ac.jp>) for details. At Christmas Island, one of those SOWER bases in the equatorial central Pacific (Figure 1), ozone and water vapor observations had been conducted in nine campaigns from 1999 to 2003. This is a very unique location since it is far from polluted air source and there has been no such observation in the equatorial central Pacific.

[7] In this study using the SOWER observation data, we will investigate mostly ozone variations at Christmas Island in the equatorial central Pacific in relation to the meteorological conditions and transport processes. Data description is in section 2. Results of ozone and water vapor variations at Christmas Island, particularly in the TTL, are presented in section 3. Section 4 summarizes our results.

2. Ozone and Water Vapor Soundings at Christmas Island

[8] For each launch at Christmas Island we used the electrochemical concentration cell (ECC) ozonesonde [Komhyr *et al.*, 1995] (EN-SCI corporation 1Z, 2Z model; 2% KI unbuffered sensor solutions) with a radiosonde (Vaisala RS80-15G, RS80-15A, and RS80-15H) to obtain a vertical distribution of ozone and meteorological parameters, such as temperature, pressure, and relative humidity. After 2000, we also used a commercial chilled-mirror hygrometer called the Snow White [Fujiwara *et al.*, 2003] with the ozonesonde. The observations at Christmas Island are summarized in Table 1. Note that we used the Vaisala ozonesonde system (the Science Pump ECC ozonesonde connected to a Vaisala GPS radiosonde) for the first campaign in 1999.

[9] Vertical distributions of ozone and meteorological parameters could be obtained from the surface to ~35 km at the maximum with a sampling interval of ~40 m. Figure 2 shows vertical profiles calculated by averaging the whole observations for ozone mixing ratio in the troposphere,

Table 1. Number of ECC Ozonesonde, Snow White Water Vapor Sonde, and Radiosonde (Vaisala RS-80) Soundings at Christmas Island (2°N, 157°W)

	Date	Ozonesonde	Water Vapor	Radiosonde
25 Feb–9 Mar 1999	28 Feb, ^a 2 Mar, ^a 4 Mar, ^a 6 Mar ^a	4 (Science Pump 6A)	–	27
17–28 Sep 1999	–	–	–	23
10–18 Mar 2000	10 Mar, 12 Mar, 14 Mar, 16 Mar, 17 Mar, 18 Mar	6 (EN-SCI 2Z)	6	6
30 Nov–6 Dec 2000	30 Nov, 1 Dec, 2 Dec, 3 Dec, 4 Dec, 5 Dec, 6 Dec	7 (EN-SCI 1Z)	7	7
27–30 Nov 2001	27 Nov, ^b 28 Nov, ^b 29 Nov, ^b 30 Nov ^b	–	4	4
19–27 Jun 2002	19 Jun, 21 Jun, 24 Jun, 25 Jun, 26 Jun, 27 Jun	6 (EN-SCI 1Z)	6	6
2 Nov 2002	2 Nov	2 (EN-SCI 1Z)	2	2
20–23 Aug 2002	20 Aug, 21 Aug, 22 Aug, 23 Aug	4 (EN-SCI 1Z)	4	4
21–24 Jan 2003	21 Jan, 22 Jan, 23 Jan, 24 Jan	4 (EN-SCI 1Z)	4	4
Total		33	33	83

^aOnly ozonesonde.

^bOnly water vapor sonde.

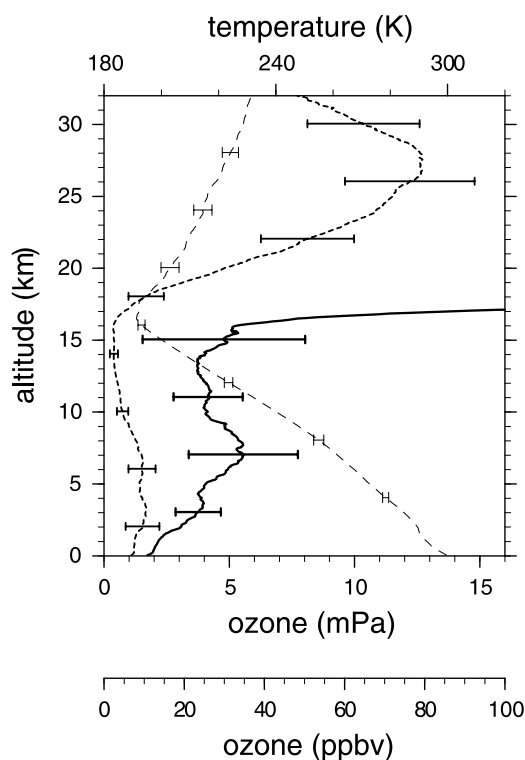


Figure 2. Mean vertical profiles of ozone volume mixing ratio (ppbv; solid line), ozone partial pressure (mPa; dashed line), and temperature (K; dotted line) at Christmas Island. Error bars indicate \pm one standard deviation.

ozone partial pressure, and temperature at Christmas Island. The cold point tropopause is found around 16.5 km in the temperature profile. The ozone partial pressure profile clearly shows the ozone layer with a maximum around 27–28 km, and a minimum just below the tropopause. Ozone mixing ratios are almost constant in the whole troposphere with reduced values in the MBL. The accuracy of the ECC ozonesonde is within ± 5 –10% [e.g., Komhyr, 1997; Oltmans et al., 2001; Smit et al., 2007], but it could be getting worse in very low ozone condition around the tropical upper troposphere. It is called the background current problem [e.g., Komhyr, 1997; Oltmans et al., 2001; Smit et al., 2007], and we need a careful treatment for such a case.

[10] In calibration for each ECC ozonesonde just before launch, we measure the background current after taking in zero ozone concentration air for about 10 min, but even in such a condition electric current occurs [Komhyr, 1997]. To derive actual ozone concentrations, we should subtract the background current from the measured electric current under an assumption that the background current is constant during each observation. Because ozone partial pressure is roughly proportional to electric current, the measurement is very sensitive to the background current in the upper troposphere where ozone partial pressure can be very low (see the minimum in Figure 2). For example in the upper troposphere, the background current error of 0.01 μ A corresponds to ~ 0.05 mPa in partial pressure and ~ 5 ppbv in mixing ratio. In the SOWER campaigns at Christmas Island except for the 1999 campaign, we carefully deter-

mined the background current for each ascent as a minimum after applying running mean (~ 39 s) to the measured current during the calibration. This timescale is reasonable, because a response time of the ECC ozonesonde is in a similar order.

[11] We used RS80 A-Humicap sensors before March 2000 and RS80 H-Humicap sensors after December 2000. About humidity observation the A-Humicap is valid up to 10 km (or -40°C) in the tropics, while the H-Humicap is valid up to 12 km (or -50°C) in relation to the temperature-dependent sensitivity of these sensors [e.g., Fujiwara et al., 2003]. In the upper troposphere, the Snow White, a commercial chilled-mirror hygrometer, has much better sensitivity and can capture small vertical variations [Fujiwara et al., 2003], although above the tropopause it cannot measure humidity correctly due to the limitation of cooling capability [Vömel et al., 2003]. Details of the Snow White including the model changes for improvement and the observations at Christmas Island are described by Fujiwara et al. [2003].

[12] We used other meteorological data sets from the European Centre for Medium-Range Weather Forecasts (ECMWF) 40-year reanalysis until August 2002 followed by the ECMWF operational analysis to cover the whole observation period. We also used the Outgoing Longwave Radiation (OLR) data from the National Oceanic and Atmospheric Administration (NOAA) to infer deep convective activity.

3. Results

3.1. Climatology in the Equatorial Central Pacific

[13] Before presenting ozonesonde observations, we briefly show the climatology of meteorological conditions in the equatorial central Pacific. We first describe the seasonal variation that is clear over the Christmas Island. Figures 3a and 3b show the 20-year climatology of the seasonal mean OLR as a proxy of deep convective activity and the seasonal mean horizontal wind at 150 hPa over the tropical Pacific. Throughout the year, an active area of large-scale convection is located around the Maritime Continent and the western Pacific. In addition, the Inter-Tropical Convergence Zone (ITCZ) extends east and west at about 5° north of Christmas Island, while the South Pacific Convergence Zone (SPCZ) extends southeastward in the southwestward of Christmas Island. That is, Christmas Island is located at rather dry area between the ITCZ and the SPCZ throughout the year, but we see some annual variations in OLR and wind fields. During northern winter (January–February–March (JFM)) convective activity along the SPCZ is enhanced (Figure 3a), while during northern summer (July–August–September (JAS)) the large-scale convective area appears over the Indochina Peninsula (Figure 3b). Meteorological fields during northern spring and autumn are basically similar to those during northern winter (not shown).

[14] Except for northern summer, the zonal wind over Christmas Island is westerly corresponding to divergence with two anticyclonic circulations being almost symmetric with respect to the equator. The anticyclonic circulations are located around the eastward of the large-scale convective area and the westward of Christmas Island (see Figure 3a at

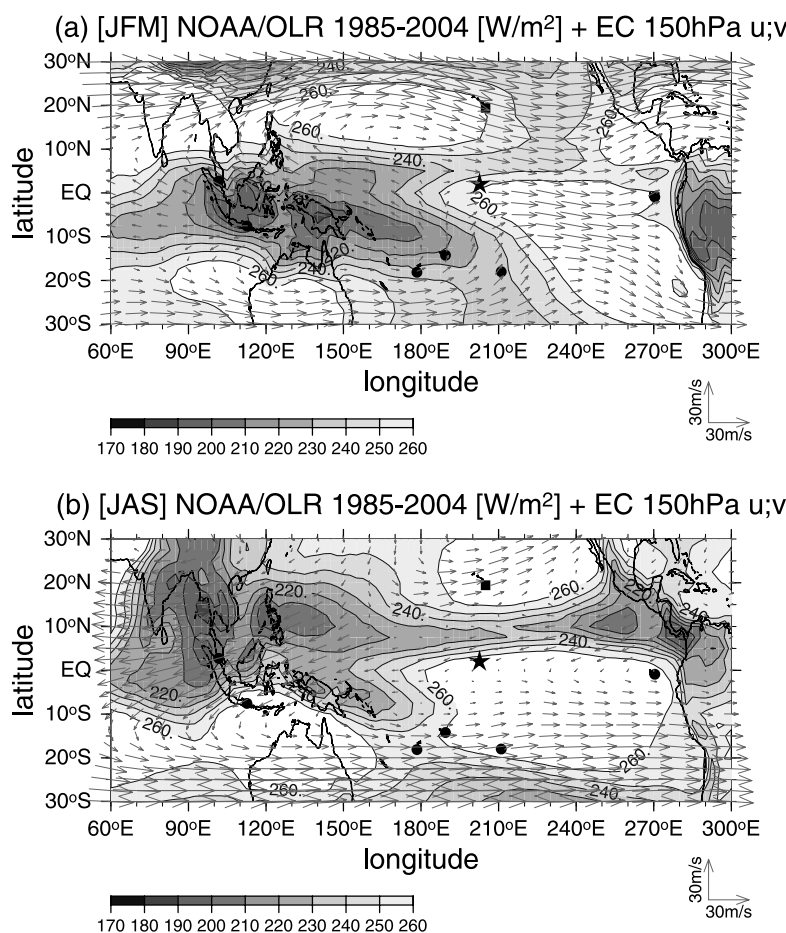


Figure 3. The 20-year mean climatology (1985–2004) of the NOAA/OLR (Wm^{-2}) and the European Centre for Medium Range Weather Forecasts (ECMWF) horizontal wind (vector) at 150 hPa for northern (a) winter (January–March) and (b) summer (July–September). The star indicates the location of Christmas Island (2°N , 157°W).

120° – 190°E , for example). During northern summer (Figure 3b) the large-scale convective area related to the Asian summer monsoon appears around the Bay of Bengal and Philippine, resulting in a weak southwestward flow over Christmas Island, while an existence of the ITCZ is clear throughout the year. The detailed observation of zonal wind at Christmas Island derived from the VHF wind profiler is described by Gage *et al.* [1996] showing the seasonal and interannual variations associated with large-scale convective activity.

[15] ENSO is a coupled ocean-atmosphere phenomenon which affects the global climate. Meteorological conditions in the equatorial central Pacific are directly affected by large-scale convection associated with the ENSO variation. Figures 4a and 4b show the OLR and horizontal wind distributions for the El Niño year and La-Niña year in northern winter, using the data for three El Niño years (1986–1987, 1991–1992, 2002–2003) and three La Niña years (1988–1989, 1998–1999, 1999–2000). In general, the convective area located in the western Pacific moves eastward in the El Niño year, bringing rather rainy condition over Christmas Island and vice versa in the La Niña year. At the same time, a divergent area with the anticyclonic circulations in the upper troposphere also moves eastward

in association with the ENSO variation. Then, the weak zonal wind is observed over Christmas Island during the El Niño phase as already reported by Gage *et al.* [1996]. The east-west movement of the meteorological fields in the equatorial Pacific is clear during northern winter (Figure 4), but the variations during northern summer is small and the fields are almost similar to the climatology (not shown). According to the southern oscillation index (SOI) prepared by the National Center for Atmospheric Research (NCAR) (<http://www.cgd.ucar.edu/cas/catalog/climind/soi.html>) for the SOWER campaign periods, we had a weak El Niño phase during 2002–2003 and weak La Niña phases during 1999–2000 and 2000–2001.

3.2. Ozone and Water Vapor Variations at Christmas Island

[16] In this subsection, we first describe characteristics of the tropospheric ozone and water vapor variations at Christmas Island, and then we investigate a relation between the ozone variations and the meteorological conditions in the upper troposphere. Figure 5 shows vertical profiles of ozone concentration and relative humidity for six campaigns out of nine to see gross features of ozone and water vapor distributions in the troposphere at Christmas Island. As seen

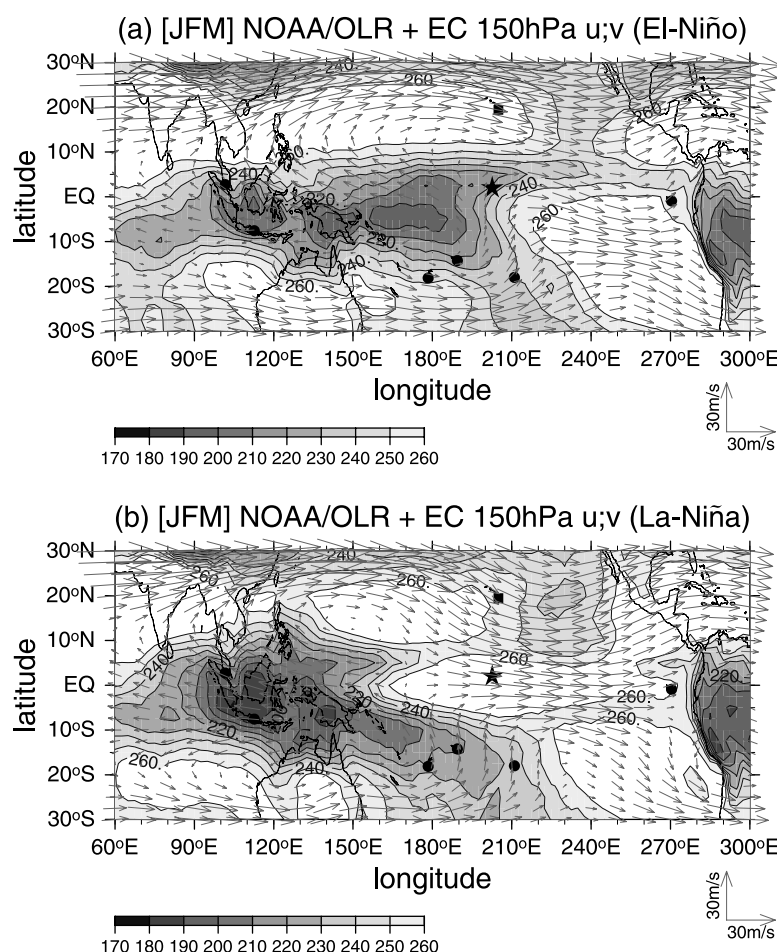


Figure 4. As in Figure 3, but for (a) the El Niño (1986–1987, 1991–1992, 2002–2003) years and for (b) the La Niña (1988–1989, 1998–1999, 1999–2000) years during January–March.

in Figure 2, the ozone mixing ratio profiles show relatively low concentrations in the whole troposphere compared with other tropical observations [e.g., Takashima and Shiotani, 2007]. We usually see a maximum in the middle troposphere and two minima in the upper troposphere and in the MBL where particularly low ozone mixing ratios with ~ 10 ppbv are found. The ozone minimum in the upper troposphere (~ 20 ppbv on the average) is located around 14 km on the mean profile, and the mixing ratio gradually increases above 14 km, possibly corresponding to the mixing barrier [Folkins *et al.*, 1999] or the bottom of the TTL, in which the convective detrainment rapid fall off above this level. However, we need to carefully investigate the processes around this level, because campaign-to-campaign variation of the level in the ozone profiles in Figure 5 is large, which means the level should be considered as a layer rather than a simple surface as discussed by Takashima and Shiotani [2007].

[17] Particularly interesting in Figure 5 are the substantially reduced ozone profiles just below the tropopause during August 2002. The mixing ratio (~ 10 ppbv at 15 km) is almost similar to that found in the MBL, and it is maintained at least during the campaign period. The substantially reduced ozone concentration in the upper troposphere such as observed during the August 2002

campaign is called “near-zero ozone concentration” that was first reported by Kley *et al.* [1996] using ozonesonde data in the western Pacific during the CEPEX campaign in March 1993. In the tropical MBL, clean (low ozone precursor gases) and wet air with ultraviolet radiation keeps the ozone mixing ratio low by photochemical reactions involving hydrogen radicals [e.g., Liu *et al.*, 1983; Routhier *et al.*, 1980; Johnson *et al.*, 1990]. The source of the near-zero ozone concentration in the upper troposphere could be due to air transport from the MBL up to the upper troposphere by deep convection [Kley *et al.*, 1996]; chemical destruction on a cirrus clouds around the tropical tropopause might be another possible factor [Roumeau *et al.*, 2000]. Our observations on the reduced ozone concentration seem to be quantitatively reasonable, because all four profiles during the campaign period show almost a similar low value with low variability. In addition, we carefully determined the background current of ~ 0.02 – $0.04 \mu A$ during the campaign period as described in section 2. Details of the near-zero ozone concentration will be described in section 3.3. The La Niña phase for the March 1999 and March 2000 campaigns may result in ozone-rich condition in the middle and upper troposphere in Figure 5, in accordance with a westward shift of convective area to the west of Christmas Island. During the January 2003

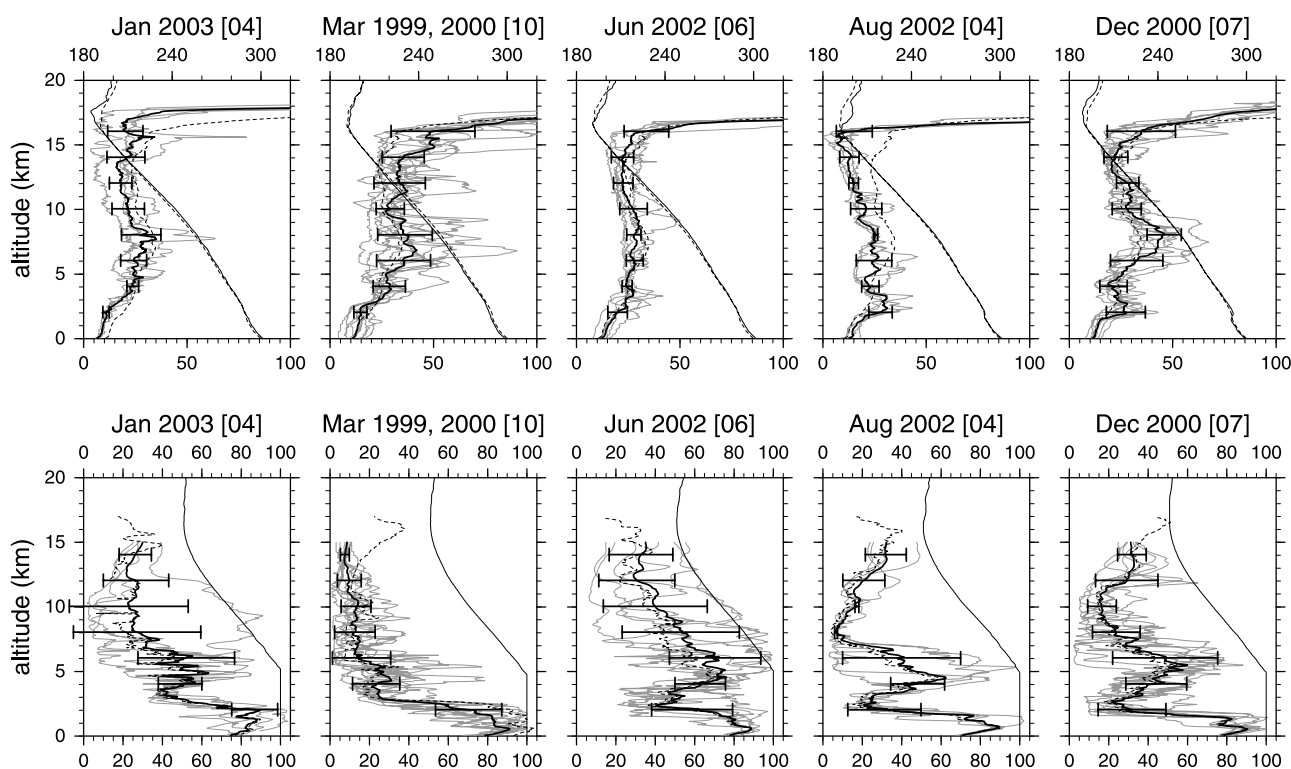


Figure 5. (top) Vertical profiles of ozone mixing ratio (ppbv) and temperature (K) and (bottom) relative humidity with respect to liquid water (%) at Christmas Island. Bold lines in the top panels indicate averaged ozone profiles, and error bars indicate \pm one standard deviation. The ozone and temperature profiles averaged over all campaigns at Christmas Island are superimposed (dashed lines). Bold lines and solid lines in the bottom panels indicate averaged relative humidity profiles and saturation relative humidity profiles, respectively (see Fujiwara et al. [2003] for details of the saturation relative humidity profiles). Averaged relative humidity profiles obtained by the Snow White are also indicated (dashed line). The number of observations for each campaign is shown in the right of the top title.

campaign in the El Niño phase, however, a convective area is very close to Christmas Island and low ozone concentrations just below the tropopause with colder tropopause temperatures (~ 185 K) are observed at Christmas Island.

[18] In the water vapor (relative humidity) profiles in Figure 5, nearly saturated air condition in the MBL (< 2 km) is persistently observed. As described above, this wet condition is important for photochemical destruction of ozone in the MBL, since the photochemical life time is less than 1 week [e.g., Kley et al., 1996]. A dry layer just above the MBL and a wet layer around 5 km are usually observed. The wet layer around 5 km is related to 0°C level clouds [e.g., Johnson et al., 1999]. Wet condition just below the tropopause is found during August 2002 campaign, when the reduced ozone is observed there. In the upper troposphere and near the tropopause, relatively dry condition was found during the March 1999 and March 2000 campaigns under the La Niña condition. In contrast, nearly saturated profile with respect to ice in the middle and upper troposphere is observed during the January 2003 campaign under the El Niño condition.

[19] Figure 6 shows the month-to-month variation of ozone concentrations at Christmas Island using all available observations, despite the small number of soundings for each month. As already described above, ozone mixing ratios are low (~ 20 ppbv) in the whole troposphere

throughout the year with substantially low ozone concentrations in the MBL. Comparing the seasonal variation at Christmas Island with other tropical stations such as Watukosek (Indonesia) and American Samoa (the equatorial Pacific) [e.g., Takashima and Shiotani, 2007], there are some differences in the seasonality. In the upper troposphere at Christmas Island the seasonal ozone minimum is found during northern summer, but at Watukosek and American Samoa it is observed during northern winter-spring. Although the seasonality is different between these observation sites, it is found to be synchronized with local convective activities. Discussion on the mechanism will be described in the following subsection.

3.3. Near-Zero Ozone Concentration in the Upper Troposphere

[20] The substantially reduced ozone concentration just below the tropopause is observed and maintained during the August 2002 campaign. Figure 7 shows the monthly averaged profiles to see how low the ozone concentration is just below the tropopause during the August 2002 campaign. The reduced ozone in the upper troposphere is first indicated by Kley et al. [1996] using the ozonesonde data in the western Pacific during the CEPEX campaign. Recently Solomon et al. [2005] investigated the spatial variation of the reduced ozone appearance in the tropics and subtropics

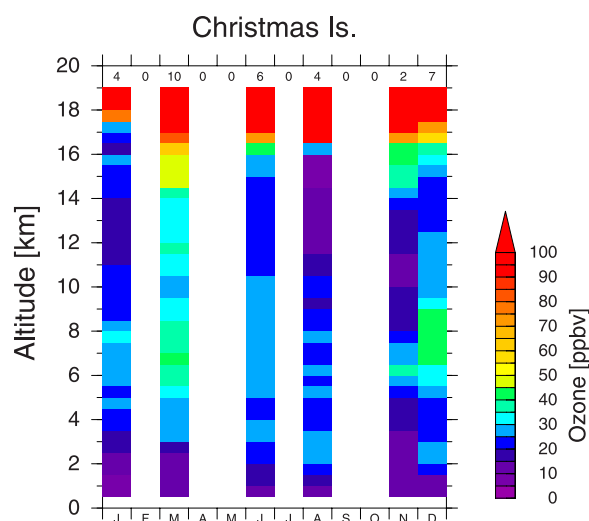


Figure 6. Vertical distributions of ozone mixing ratio (ppbv) for each month averaged in 0.5-km bins at Christmas Island. The number of observations for each month is shown in the upper part of this figure.

using the SHADOZ archive, and found that high frequency of the reduced ozone appearance is observed in the equatorial southern Pacific, such as at American Samoa and also at Watukosek. The maximum fraction is observed at ~ 200 hPa, and it is getting small as close to the tropopause [see Solomon *et al.*, 2005, Figure 3].

[21] Since Solomon *et al.* [2005] did not show the seasonality of reduced ozone appearance in the tropics, in Figure 8 we investigate vertical profiles of the seasonal frequency at Watukosek (Indonesia) and American Samoa (the equatorial southern Pacific) where the larger values are obtained by Solomon *et al.* [2005]. Here the frequency is defined as a rate lower than 20 ppbv, which is used by Solomon *et al.* [2005]. In addition to the fraction maxima in the upper troposphere similar to those shown by Solomon *et al.* [2005], we can clearly see the seasonal variation in the upper troposphere with maxima mostly during northern winter and spring at both sites. This is consistent with the seasonal variation of convective activities in the tropics as seen in Figure 3. During northern winter-spring convective activities over the equator and the SPCZ covering Watukosek and American Samoa are enhanced. That is, seasonality of the fraction in the upper troposphere is supposed to be due to vertical transport of low-ozone air in the MBL by deep convection. In the equatorial southern Pacific (American Samoa), an altitude of the maximum fraction is almost constant along the seasonal variation and the profiles shift in parallel. The seasonal variation of the ozone concentration around the altitude is partly related to the seasonal variation of ozone in the MBL with maxima during northern summer as shown by Oltmans and Komhyr [1986]. On the other hand in Indonesia (Watukosek), there is a seasonal change in the maximum altitude, suggesting a relation to the seasonality in convective activity (see around 14–16 km), although there is a high fraction during northern summer around 10–12 km. As already indicated by Solomon *et al.* [2005], the fraction near the surface at Watukosek is quite low, and the high fractions in the upper

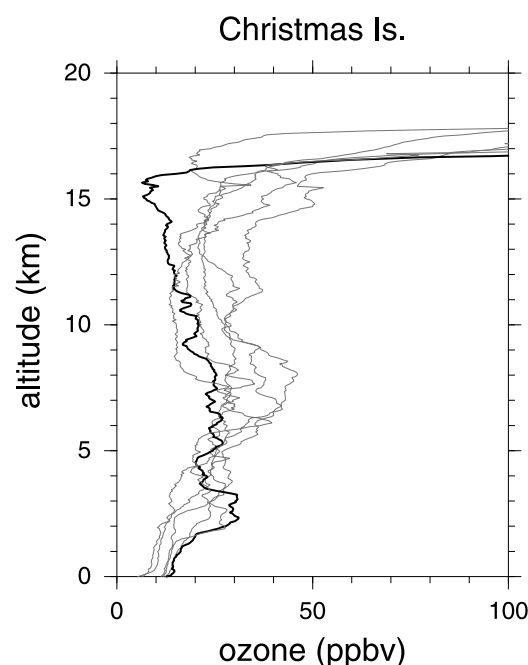


Figure 7. Monthly mean profiles of ozone mixing ratio (ppbv) at Christmas Island. The bold line shows the ozone profile for August 2002.

troposphere could be due to advection of low-ozone air from the ocean area. The low fraction at Watukosek near surface is due to air pollution.

[22] Figure 9 shows a map of the OLR and horizontal wind fields at 150 hPa during the August 2002 campaign. The large-scale convective area is observed over the Bay of Bengal and the western Pacific, and the wind field in the upper troposphere is a weak southwestward flow over Christmas Island, suggesting advection from the ITCZ, though the maximum convective area in the western Pacific is found slightly westward ($\sim 170^\circ\text{E}$) from the climatology in Figure 3. The source of reduced ozone air over Christmas Island seems to be the MBL origin in the ITCZ. From the trajectory analysis air around this altitude was advected

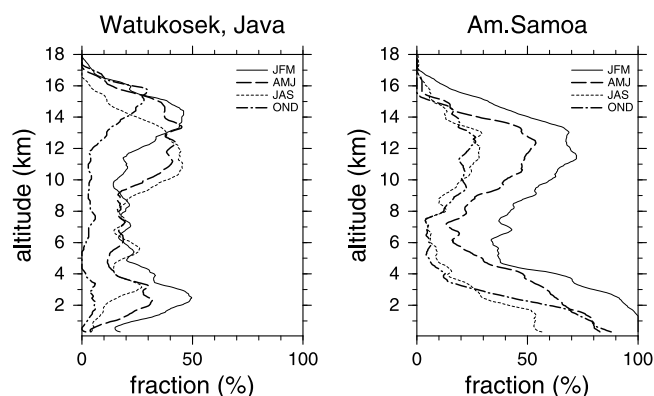


Figure 8. Vertical profiles of the occurrence frequency of ozone mixing ratios below 20 ppbv at Watukosek (Indonesia) and American Samoa for each season. The analysis is made for the period using mid-1998 to mid-2003 from the SHADOZ archive.

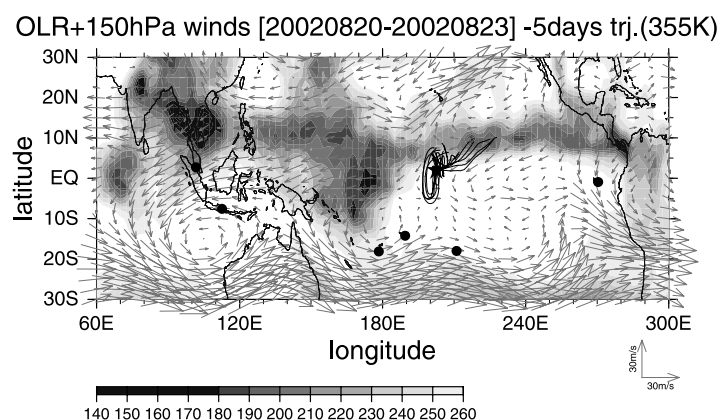


Figure 9. A map of the OLR (Wm^{-2}) and the horizontal wind fields at 150 hPa during the August 2002 campaign (4-day mean). Five-day isentropic back-trajectories on 355 K level (~ 15 km) starting every 6 hours from 0000 UTC 20 August 2002 to 0000 UTC 24 August 2002 are also superimposed. The trajectory calculation using horizontal wind fields from the ECMWF 40-year reanalysis is stopped when trajectory passes over a deep convective area ($\text{OLR} < 190 \text{ Wm}^{-2}$). The star indicates the location of Christmas Island (2°N , 157°W).

from the ITCZ ~ 2 – 4 days ago, although we may need further consideration of the trajectory calculation with the reanalysis data in the TTL. The timescale of advection can be short enough to keep the low ozone mixing ratio from the ITCZ. It is important to recall the annual variation of the horizontal wind over Christmas Island with a weak southwestward flow during northern summer (Figure 3). During the CEPEX campaign, there were deep convective clouds in the SPCZ very close to the observation point in some cases [Kley *et al.*, 1996], a distance from deep convective cloud may be another important factor for the transport process in the tropical upper troposphere.

[23] Low-ozone air mass in the MBL can be supplied to the upper troposphere by deep convection in the ITCZ throughout the year, but the air can be advected to Christmas Island only in northern summer in accordance with the southwestward wind field. This is controlled by a large-scale convective area usually located in the western Pacific, and may also be affected by a convective area associated with the American summer monsoon that is found during northern summer to the northeast of Christmas Island. The different seasonality between Christmas Island and other Pacific stations is due to a seasonal change in its location of large-scale convection and associated large-scale airflow in the upper troposphere.

4. Summary

[24] We have conducted ozone and water vapor observation campaigns at Christmas Island (2°N , 157°W ; Figure 1) in the equatorial central Pacific as a part of the SOWER/Pacific mission. From 1999 to 2003, nine campaigns have been performed with 33 ozonesondes (ECC) and 33 chilled-mirror hygrometers (Snow White) as shown in Table 1. This is one of the most important regions in the equatorial Pacific to consider the east-west contrast of meteorological conditions associated with the Walker circulation and its variation due to ENSO.

[25] From ozonesonde observations, we found that ozone concentration at Christmas Island is low over the whole

troposphere. In particular just below the tropopause during the August 2002 campaign period we observed substantially reduced ozone concentrations (< 10 ppbv) similar to those found in the MBL, which are maintained at least during the campaign period. The observed seasonality of the reduced ozone concentration just below the tropopause at Christmas Island differ from those at Watukosek (Indonesia) and at American Samoa (the equatorial southern Pacific) where Solomon *et al.* [2005] observed the maximum appearance during northern winter and spring in the upper troposphere. From meteorological conditions we found that air was advected from the ITCZ located at 300 – 1000 km north of Christmas Island, in accordance with the northeasterly wind in the upper troposphere, suggesting that the origin of air mass is from the MBL in the ITCZ.

[26] Although measurements on the substantially reduced ozone concentration can be quantitatively reasonable with our careful treatment of the ECC calibration, additional observations may be needed for more detailed discussion. For future studies on the mechanism of the reduced ozone concentration in the TTL, a quantitative estimate on chemical processes, such as chemical reaction on ice as indicated by Roumeau *et al.* [2000] may be important. We need additional investigation for various processes in the TTL involving ozone variation to clarify the STE mechanisms.

[27] **Acknowledgments.** We would like to thank staff members of MiniHotel at Christmas Island for logistic support. The authors thank H. Hayashi of Kyoto University. We also thank anonymous reviewers for constructive comments on the article. We thank the SHADOZ project for providing ozonesonde data in the tropics and the ECMWF for meteorological analysis and reanalysis data. The figures were produced with the GFD-Dennou Library. This study was supported by the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT) through the Grant-in-Aid for Scientific Research (11219201, 15204043, and 18204041) and by the Ministry of the Environment through the Global Environment Research Fund (A-1 and A-071).

References

Fishman, J., C. E. Watson, J. C. Larsen, and J. A. Logan (1990), Distribution of tropospheric ozone determined from satellite data, *J. Geophys. Res.*, **95**, 3599–3617.

- Folkins, I., M. Loewenstein, J. Podolske, S. J. Oltmans, and M. Proffitt (1999), A barrier to vertical mixing at 14 km in the tropics: Evidence from ozonesondes and aircraft measurements, *J. Geophys. Res.*, **104**, 22,095–22,102.
- Folkins, I., P. Bernath, C. Boone, G. Lesins, N. Livesey, A. M. Thompson, K. Walker, and J. C. Witte (2006), Seasonal cycles of O₃, CO, and convective outflow at the tropical tropopause, *Geophys. Res. Lett.*, **33**, L16802, doi:10.1029/2006GL026602.
- Fujiwara, M., M. Shiotani, F. Hasebe, H. Vömel, S. J. Oltmans, P. W. Ruppert, T. Horinouchi, and T. Tsuda (2003), Performance of the Meteorolabor “Snow White” chilled-mirror hygrometer in the tropical troposphere: Comparisons with the vaisala RS80 A/H-Humicap sensors, *J. Atmos. Oceanic Technol.*, **20**, 1534–1542.
- Gage, K., J. McAfee, and C. Williams (1996), On the annual variation of tropospheric zonal winds observed above Christmas Island in the central equatorial Pacific, *J. Geophys. Res.*, **101**, 15,061–15,070.
- Hasebe, F., M. Fujiwara, N. Nishi, M. S. H. Voemel, S. Oltmans, H. Takashima, S. Saraspriya, N. Komala, and Y. Inai (2007), In situ observations of dehydrated air parcels advected horizontally in the tropical tropopause layer of the western Pacific, *Atmos. Chem. Phys.*, **7**, 803–813.
- Johnson, J. E., R. H. Gammon, J. Larsen, T. S. Bates, S. J. Oltmans, and J. C. Farmer (1990), Ozone in the marine boundary layer over the Pacific and Indian Oceans: Latitudinal gradients and diurnal cycles, *J. Geophys. Res.*, **95**, 11,847–11,856.
- Johnson, R. H., T. M. Rickenbach, S. A. Rutledge, P. E. Ciesielski, and W. H. Schubert (1999), Trimodal characteristics of tropical convection, *J. Clim.*, **12**, 2397–2418.
- Kley, D., P. J. Crutzen, H. G. J. Smit, H. Vömel, S. J. Oltmans, H. Grassl, and V. Ramanathan (1996), Observations of near-zero ozone concentrations over the convective Pacific: Effects on air chemistry, *Science*, **274**, 230–233.
- Kley, D., H. G. J. Smit, H. Vömel, H. Grassl, V. Ramanathan, P. J. Crutzen, S. Williams, J. Meywerk, and S. J. Oltmans (1997), Tropospheric water vapor and ozone cross-sections in a zonal plane over the central equatorial Pacific Ocean, *Q. J. R. Meteorol. Soc.*, **123**, 2009–2040.
- Kley, D., J. M. Russell III, and C. Phillips (Eds.) (2000), SPARC assessment of upper tropospheric and lower stratospheric water vapor, *Rep. WCRP 113, WMO/TD-No 1043*, World Meteorol. Org., Geneva, Switzerland.
- Komhyr, W. D. (1997), Model KZ-ECC atmospheric ozone sounding system, report, EN-SCI Corporation, Boulder, Colo.
- Komhyr, W. D., R. A. Barnes, G. B. Brothers, J. A. Lathrop, and D. P. Opperman (1995), Electrochemical concentration cell ozonesonde performance evaluation during STOIC 1989, *J. Geophys. Res.*, **100**, 9231–9244.
- Liu, S. C., M. McFarland, D. Kley, O. Zafiriou, and B. Huebert (1983), Tropospheric NO_x and O₃ budgets in the equatorial Pacific, *J. Geophys. Res.*, **88**, 1360–1368.
- Oltmans, S. J., and W. D. Komhyr (1986), Surface ozone distributions and variations from 1973–1984 measurements at the NOAA geophysical monitoring for climatic-change base-line observatories, *J. Geophys. Res.*, **91**, 5229–5236.
- Oltmans, S. J., et al. (2001), Ozone in the Pacific tropical troposphere from ozonesonde observations, *J. Geophys. Res.*, **106**, 32,503–32,526.
- Roumeau, S., P. Brémaud, E. Rivière, S. Baldy, and J. L. Baray (2000), Tropical cirrus clouds: A possible sink for ozone, *Geophys. Res. Lett.*, **27**, 2233–2236.
- Routhier, F., R. Dennett, D. D. Davis, A. Wartburg, P. Haagensohn, and A. C. Delany (1980), Free tropospheric and boundary-layer airborne measurements of ozone over the latitude range of 58 S to 70 N, *J. Geophys. Res.*, **85**, 7307–7321.
- Shiotani, M. (1992), Annual, quasi-biennial, and El Niño–Southern Oscillation (ENSO) time-scale variations in equatorial total ozone, *J. Geophys. Res.*, **97**, 7625–7633.
- Smit, H. G. J., et al. (2007), Assessment of the performance of ECC-ozonesondes under quasi-flight conditions in the environmental simulation chamber: Insights from the Juelich Ozone Sonde Intercomparison Experiment (JOSIE), *J. Geophys. Res.*, **112**, D19306, doi:10.1029/2006JD007308.
- Solomon, S., D. W. J. Thompson, R. W. Portmann, S. J. Oltmans, and A. M. Thompson (2005), On the distribution and variability of ozone in the tropical upper troposphere: Implications for tropical deep convection and chemical-dynamical coupling, *Geophys. Res. Lett.*, **32**, L23813, doi:10.1029/2005GL024323.
- Takashima, H., and M. Shiotani (2007), Ozone variation in the tropical tropopause layer as seen from ozonesonde data, *J. Geophys. Res.*, **112**, D11123, doi:10.1029/2006JD008322.
- Thompson, A. M., et al. (2003), Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology: 2. Tropospheric variability and the zonal wave-one, *J. Geophys. Res.*, **108**(D2), 8241, doi:10.1029/2002JD002241.
- Vömel, H., S. J. Oltmans, D. Kley, and P. J. Crutzen (1995), New evidence for the stratospheric dehydration mechanism in the equatorial Pacific, *Geophys. Res. Lett.*, **22**, 3235–3238.
- Vömel, H., M. Fujiwara, M. Shiotani, F. Hasebe, S. J. Oltmans, and J. E. Barnes (2003), The behavior of the Snow White chilled-mirror hygrometer in extremely dry conditions, *J. Atmos. Oceanic Technol.*, **20**, 1560–1567.
- M. Fujiwara and F. Hasebe, Faculty of Environmental Earth Science, Hokkaido University, Sapporo, 060-0810, Japan.
- N. Nishi, Geophysical Institute, Kyoto University, Kyoto, 606-8502, Japan.
- M. Shiotani, Research Institute for Sustainable Humanosphere, Kyoto University, Uji, Kyoto, 611-0011, Japan. (hisahiro@rish.kyoto-u.ac.jp)
- H. Takashima, Atmospheric Composition Research Program, Frontier Research Center for Global Change, JAMSTEC, 3173-25 Showa-machi, Kanazawa-ku, Yokohama, Kanagawa 236-0001, Japan. (hisahiro@jamstec.go.jp)